

DECAY OF  $\text{Tm}^{174}$ 

S. C. GUJRATHI AND S. K. MUKHERJEE

SAHA INSTITUTE OF NUCLEAR PHYSICS, CALCUTTA, INDIA.

(Received February 16, 1967)

**ABSTRACT.** The  $\text{Tm}^{174}$  nucleus is produced by  $\text{Yb}^{174}(n, p)\text{Tm}^{174}$  reaction with 14.8-MeV neutrons on enriched (99%) Yb isotope and is studied by means of beta-gamma scintillation counter techniques. The beta and gamma measurements show beta groups of end-point energies  $1200 \pm 50$  (82%) and  $700 \pm 50$  (18%) keV and gamma rays of energies 50 (Yb K X-ray), 75, 175, 275, 350, 370, 500, 630, 870, 990, 1260 and 1350 keV decaying with a  $5.5 \pm 0.5$ -min activity of  $\text{Tm}^{174}$ . The gamma-gamma coincidence studies confirm previously reported gamma-ray cascades along with a new cascade of 500-1350-275-75 keV. The gamma spectrum in coincidence with the beta rays above 300 keV shows the existence of the previously unreported 1350-keV gamma transition in the new cascade and also gives the 870-keV gamma transition between the 2380-keV level and the 1510-keV isomeric state of  $\text{Yb}^{174}$ . A decay scheme of  $\text{Tm}^{174}$  is proposed and the results are compared and discussed in the light of the unified model and the pairing correlation model. The  $(n, p)$  cross-section for  $\text{Yb}^{174}$  is found to be  $3.5 \pm 1$  mb.

## INTRODUCTION

Wille and Fink (1960) reported for the first time the production of the nuclide  $\text{Tm}^{174}$  by neutron irradiations of enriched  $\text{Yb}^{174}$  and assigned a 5.5-min half-life to it. Takahashi *et al* (1961) observed a 5-min activity in the products formed by the fast neutron bombardment on natural ytterbium oxide. Kantele *et al* (1964) made a study of the  $\text{Tm}^{174}$  isotope and reported its half-life as  $5.2 \pm 0.3$  min after following the decay of the 1000-keV gamma ray in a single-channel analyser. They also reported a decay scheme of  $\text{Tm}^{174}$  based on the results of sum and sum-peak spectra along with the knowledge of the 850- $\mu\text{s}$  isomeric state (Kantele, 1964) at 1520 keV in  $\text{Yb}^{174}$ . According to the decay scheme of Kantele *et al.* (1964), the  $\text{Tm}^{174}$  nucleus decays by two beta groups of maximum energies  $1200 \pm 100$  ( $\approx 80\%$ ) and 0.700 keV ( $\approx 20\%$ ) to the excited levels of  $\text{Yb}^{174}$  at 1886 and 2386 keV, respectively. The 2385-keV level decays by the 497-keV gamma transition to the 1886-keV level, which in turn populates the 850- $\mu\text{s}$  isomeric state at 1520 keV. The isomeric state apparently depopulates by a multipole cascade of the 995-275-175-76 keV gamma rays. They have also tentatively suggested other weak modes of decay of the isomeric state through the cascades 1270-175-76 keV and 630-366-275-76 keV. It is known (Orth, 1964) from the decay of the 165-d isomer of  $\text{Lu}^{174}$  that the isomeric state at 1520-keV in  $\text{Yb}^{174}$  decays to  $4^+$ ,  $6^+$ , and  $8^+$  members of the ground state rotational band with relative intensities  $\approx 0.1$ , 1.0 and  $\approx 0.01$ . Very recently Funke *et al* (1965) in their study of  $\text{Lu}^{174m}$  reported the decay of the 1526-keV isomeric state

of  $\text{Yb}^{174}$  in two cascades 995-280-175-76.5 and 630-365-280-175-76.5 keV having the intensity ratio  $41 \pm 5$ . They have not observed the third cascade 1270-175-76.5 keV reported by previous authors (Kantele, 1964; Orth, 1964).

The present investigation was undertaken to study the decay of  $\text{Tm}^{174}$  in more detail with an intention of studying the weak gamma transitions assigned tentatively by Kantele *et al.* (1964) and also to resolve the discrepancies regarding the number of cascades and their relative intensities observed in the de-excitation of the isomeric state of  $\text{Yb}^{174}$ .

#### SOURCE PREPARATION

The  $\text{Tm}^{174}$  isotope was produced by the  $(n, p)$  reaction on  $\text{Yb}^{174}$  enriched to 99%. The incident neutron energy was 14.8 MeV, and the typical neutron flux of the order of  $4 \times 10^{10}$  n/cm<sup>2</sup> sec was maintained at this level in all the irradiations. The possible contaminant activities in the source were carefully looked for on the basis of the data of isotopic and spectrographic analysis of the supplied enriched sample as well as with the aid of suitable activity lists (Slater, 1962; Nuclear Data Sheets, 1964). A long-lived activity noticed as an impurity in a very small amount was due to  $\text{Yb}^{176}$  (4.1 d). This was produced by the  $(n, 2n)$  reaction on  $\text{Yb}^{176}$  which was present as much as 0.4% in the enriched isotope. The contribution due to this activity was subtracted out in all the measurements. The other short-lived activities, suspected as impurities, were due to the reactions  $\text{Cu}^{63} (n, 2n) \text{Cu}^{62}$  (9.5 min);  $\text{Pr}^{141} (n, 2n) \text{Pr}^{140}$  (3.4 min) and  $\text{Yb}^{176} (n, p) \text{Tm}^{176}$  (1.4 min). The contribution of  $\text{Tm}^{176}$  is expected to be very small since the measured  $(n, p)$  cross-section for  $\text{Yb}^{176}$  is  $1.5 \pm 0.5$  mb (Gujrathi *et al.*, 1965). The impurities of  $\text{Cu}^{63}$  and  $\text{Pr}^{141}$  present in the enriched sample were insufficient to alter the main conclusions. The estimated total contribution due to all these impurities was less than 3% of the total decay of  $\text{Tm}^{174}$ .

For the beta ray measurements the irradiated enriched ytterbium in the form of oxide powder was uniformly spread inside a bag of thin mylar. The average thickness of the source was 1 mg/cm<sup>2</sup>. Since the measured  $(n, p)$  cross-section for  $\text{Yb}^{174}$  is small, a large number of irradiations on several hundred milligrams of enriched isotopes were needed to complete each measurement.

#### EXPERIMENTAL RESULTS

##### (A) Half-life measurement and the $(n, p)$ cross-section

The half-life of  $\text{Tm}^{174}$  was measured as  $5.5 \pm 0.3$  min in close agreement with previous reports. The half-life was also studied by following the decay of 50, 175, 275, 370 and 990-keV gamma rays (see Table 1) in a single-channel analyser. In all cases a clear half-life of  $5 \pm 1$  min was observed. The cross-section for the  $\text{Yb}^{174} (n, p) \text{Tm}^{174}$  reaction was calculated by the activation technique, the

## Decay of $Tm^{174}$

experimental details of which were as reported earlier (Mukherjee *et al.*, 1961). The measured value of the cross-section was  $3.5 \pm 1$  mb as compared with the value of 556 mb for the  $Cu^{63} (n, 2n) Cu^{62}$  reaction.

### (B) Gamma and beta ray measurements

The experiments were performed to study the gross features of the gamma ray spectra of  $Tm^{174}$  produced by bombarding enriched  $Yb^{174}$ . The gamma spectra were measured with a  $7.6 \text{ cm} \times 7.6 \text{ cm}$  NaI(Tl) crystal optically coupled to a RCA-8054 photomultiplier and a 512-channel analyser. Fig. 1 shows a typical gamma ray spectrum in which the source was placed at a distance of 3 cm from the crystal on a 0.5 cm thick lucite plate to cut off the beta particles. Photo peaks are observed at energies 50, 75, 175, 275, 370, 490, 630, 870, 990, 1260 and 1340 keV. Due to the summing of the cascade gamma rays some of the peaks have been slightly broadened. This effect is clearly seen in the 490 and 990-keV gamma rays. The gamma spectra were also studied by systematically introducing 0.1 to 1 cm thick lead absorbers between the source and the detector to reduce the summing of X-rays and the low energy gamma rays. The broadenings at 550 keV and 1040 keV shown in fig. 1, were found to be due to the Yb K X-ray or the low-energy gamma ray summing with the 490 and 990 keV gamma rays. To study the decay characteristics of the photopeaks observed in the single-crystal measurements, another experiment was performed in which three gamma spectra were taken in succession at 1 min, 5 min and 11 min after the end of the

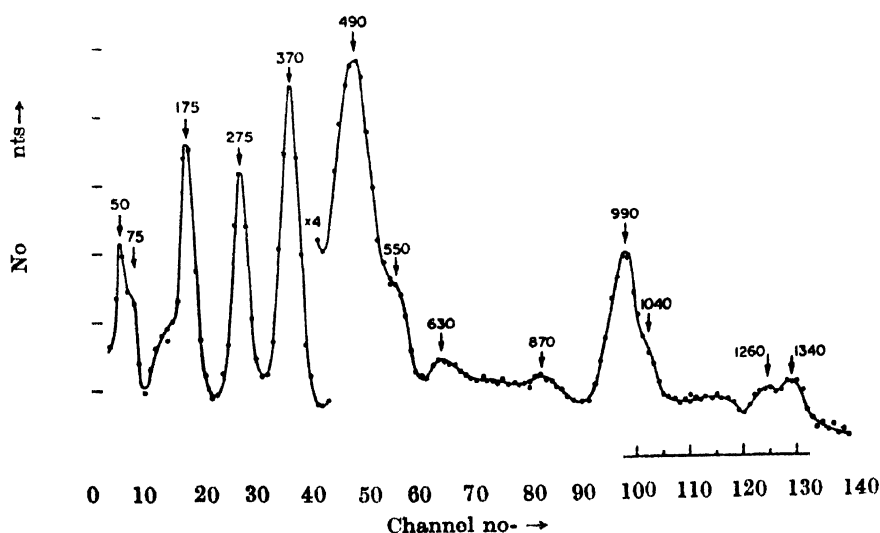


Fig. 1. Gamma spectrum of  $Tm^{174}$  taken with a  $7.6 \text{ cm} \times 7.6 \text{ cm}$  NaI(Tl) detector. Source-to-crystal distance was 3 cm.

bombardment. Fig. 2 shows that all the prominent gamma rays are decaying with a half-life of  $5 \pm 1$  min.

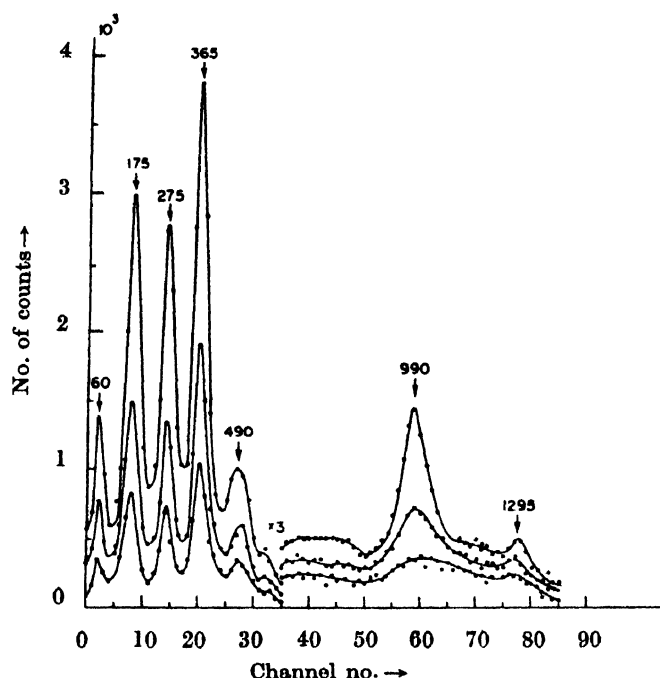


Fig. 2. Gamma spectra of  $\text{Tm}^{174}$  taken at 1 min, 6 min, and 11 min. after the end of the irradiation of enriched  $\text{Yb}^{174}$  shown as curves (a), (b) and (c), respectively.

The gamma ray energies, their relative intensities calculated from single crystal as well as from some of the coincidence measurements and the gamma-gamma coincidence results are given in table 1.

TABLE 1

Gamma ray energies, their relative intensities neglecting the conversions and the gamma-gamma coincidence results.

Energy of the photo peak (keV)	Unconverted gamma-ray relative intensity	In coincidence with (keV)
$50 \pm 5$ (Yb K X-ray)	$30 \pm 10$	175, 275, 350, 500, 630, 990, 1260, 1350
$75 \pm 5$	$10 \pm 3$	
$175 \pm 5$	$72 \pm 15$	50, 275, 350, 500, 630, 990, 1260, 1350
$275 \pm 5$	$90 \pm 15$	50, 175, 350, 500, 630, 990, 1350
$350 \pm 10$	$7 \pm 2$	50, 175, 275, 500, 630
$370 \pm 10$	100	
$500 \pm 10$	$15 \pm 4$	370
$630 \pm 10$	$5 \pm 2$	
$870 \pm 10$	$5 \pm 2$	
$990 \pm 10$	$75 \pm 15$	50, 175, 275
$1260 \pm 15$	$6 \pm 2$	50, 175
$1350 \pm 15$	$7 \pm 2$	50, 175, 275

The beta measurements were performed with a 5.1 cm  $\times$  1.3 cm thick anthracene crystal mounted on a RCA-6810A photomultiplier and a 512-channel analyser. The source to crystal distance was 0.5 cm. Fig. 3 shows the Fermi-Kurie analysis of the beta spectrum of  $Tm^{174}$ . Two beta groups with maximum energies of  $1200 \pm 50$  (82%) and  $700 \pm 50$  (18%) keV are found associated with the decay of  $Tm^{174}$ . The energies, intensities and the log ft values of the two

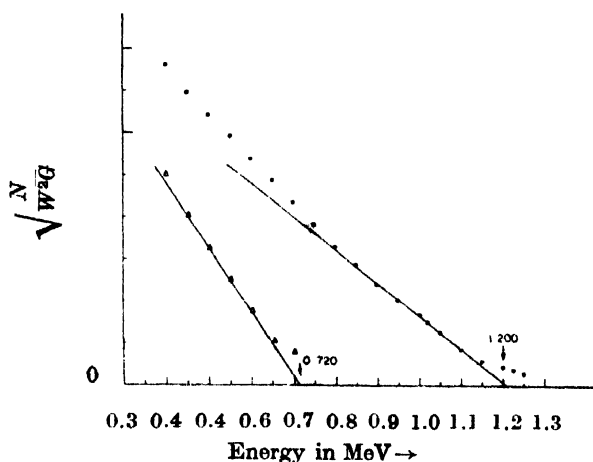


Fig. 3. Fermi-Kurie plot of the beta spectrum of  $Tm^{174}$ . The detector was a 5.1 cm  $\times$  1.3 cm. thick anthracene crystal. Source-to-detector distance was 0.5 cm.

beta groups are given in table 2. In the measurement a very weak beta group of  $2500 \pm 400$  keV (2-3%) was observed to decay with  $6 \pm 2$  min-activity. This group was not assigned to  $Tm^{174}$  as it was found in coincidence with the 510-keV annihilation gamma rays. It might be due to the suspected impurities of  $Cu^{62}$  (9.5 min) and  $Pr^{140}$  (3.5 min) present in very small amounts in the  $Tm^{174}$  sources

TABLE 2

Energies, intensities and log ft values of the beta ray groups.

Beta ray end-point energy (keV)	Intensity (%)	log ft. value
$1200 \pm 50$	82	4.8
$700 \pm 50$	18	4.6

## DISCUSSION

### (C) Coincidence results

To study the cascade nature of the various gamma rays and beta-gamma relationships, coincidence studies were made. In the gamma-gamma coincidence studies, a 5.1 cm  $\times$  5.1 cm NaI(Tl) crystal spectrometer was used to select the

gating gamma ray and the gated spectrum was taken with another 7.6 cm  $\times$  7.6 cm NaI(Tl) crystal. For the beta-gamma coincidence measurements the 7.6 cm  $\times$  7.6 cm NaI(Tl) in the above arrangement was replaced by a 5.1 cm  $\times$  1.3 cm thick anthracene crystal. The resolving time ( $2\tau$ ) of the coincidence system was 2  $\mu$ s.

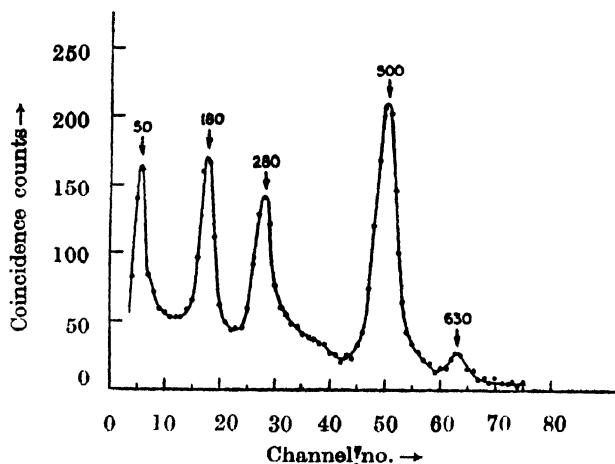


Fig. 4. Gamma spectrum in coincidence with the 370-keV gamma ray.

Most of the prominent gamma rays observed in the single-crystal spectrum were taken in the gate, and the coincidence gamma spectra were studied. The spectrum in coincidence with the Yb K X-ray showed clear photo peaks at 175, 275, 350, 500, 630, 990, 1260 and 1350 keV. The existence of the fourfold cascade of an X-ray and 175, 275 and 990-keV gamma rays reported by Kantele *et al.*, was confirmed. With the 370-keV gamma ray in the gate the 500-keV gamma ray was observed in coincidence with the other photopeaks at 50, 180, 280 and 630 keV as shown in fig. 4. With the 275-keV gamma ray in the gate, the coincidence spectrum showed the 55, 75, 175, 275, 350, 500, 635, 990 and 1350-keV gamma rays (fig. 5); while with the 175-keV gamma ray all these gamma rays with an addition of the 1260-keV gamma ray appeared in coincidence. In fig. 6, curve (a) shows the result obtained by gating with the 1200-1400-keV portion of the gamma-ray spectrum, which includes both the photopeaks of energies 1260 and 1340 keV and curve (b) shows the results on gating with the 1200-1260-keV portion of the gamma spectrum which includes mostly the 1260-keV photopeak. It can be seen from fig. 6 that the intensities of the 275 and 500-keV photopeaks are decreased in curve (b) relative to the intensities of the 50 and 175-keV gamma rays. All the gamma-gamma coincidence results are summarised in table 1.

The beta spectrum in coincidence with the 370-keV gamma ray showed two beta groups of end point energies  $1200 \pm 50$  and  $700 \pm 50$  keV with the same intensities as observed in the single-crystal measurement. When the beta spec-

spectrum was gated with the 500-keV gamma ray, a single beta group of  $700 \pm 50$  keV was observed in coincidence. With the 990-keV gamma rays no beta particles

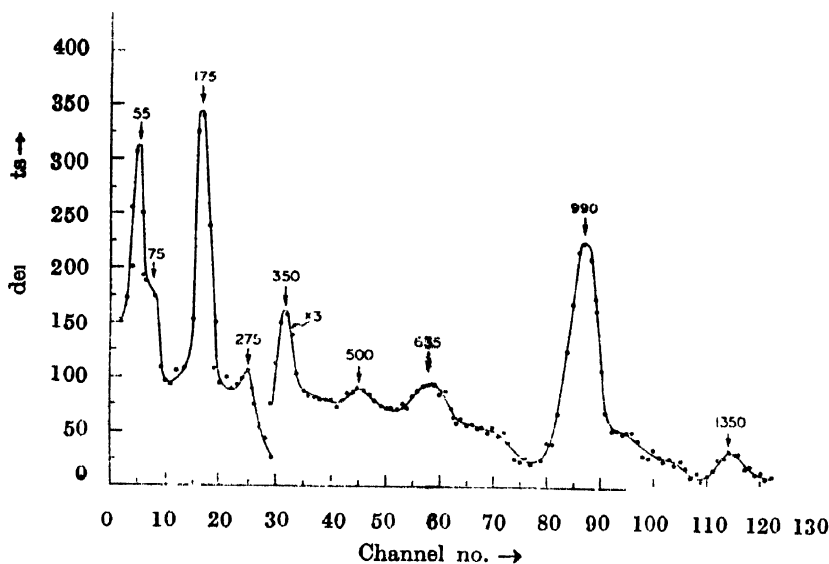


Fig. 5. Gamma spectrum in coincidence with the 275-keV gamma ray.

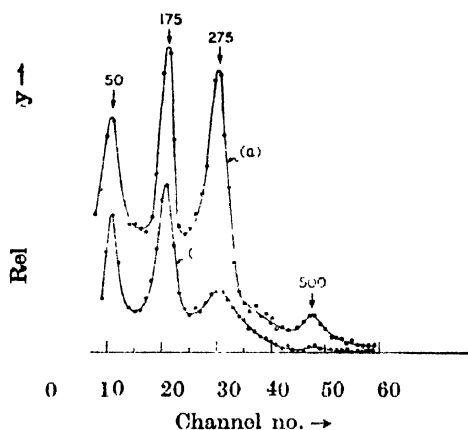


Fig. 6. Gamma spectra obtained in coincidence when the gating window was set at the 1200-1400 and 1200-1260-keV portions of the gamma spectrum, shown as curves (a) and (b), respectively.

were observed in prompt coincidence. The coincidence spectra were also studied by gating with the 275 and 175-keV gamma rays which showed an indication of the 1200-keV beta group. The results were not conclusive as it was difficult to estimate the contributions due to the spectra obtained by beta particles and the Compton scattered electrons from the coincident high-energy gamma rays. To cross-check this result and also to find gamma rays in coincidence with the beta particles, a gamma spectrum in coincidence with the beta rays was studied.

The gated beta spectrum was biased up to 300 keV to avoid the contribution of high energy gamma rays arising due to gamma-gamma coincidences with the low-energy ones. Fig. 7 shows this coincidence result with the evidence for the existence of the photo peaks at 50, 175, 275, 370, 500, 870 and 1350 keV.

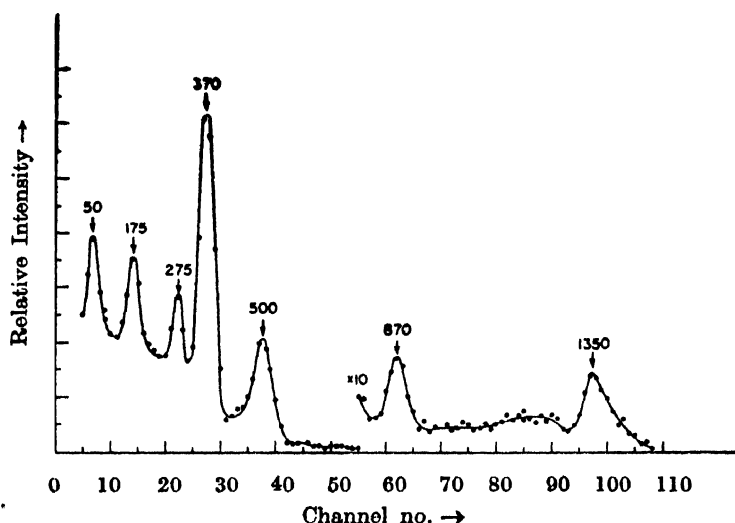


Fig. 7. Gamma spectrum in coincidence with the beta rays greater than 300 keV.

In the spectrum the intensities of the low-energy gamma rays of energies 50, 175 and 275 keV is slightly higher due to the coincidences between these gamma rays and the high energy coincident gamma rays detected in anthracene crystal.

#### DISCUSSION

The  $(n, p)$  cross-section value for  $\text{Yb}^{174}$  is  $3.5 \pm 1$  mb and is comparable with the observed  $(n, p)$  cross sections in this mass region of rare earth (Wille *et al.* 1960; Gardner, 1962; Chatterjee, 1964). This value is higher than the one predicted by Gardner (1962) which is 2 mb. It also fits quite satisfactorily with the trends of  $(n, p)$  reaction cross-sections at 14-MeV neutrons (Chatterjee, 1964).

A proposed decay scheme of  $\text{Tm}^{174}$  which contains all the prominent features of the observations is shown in fig. 8. This agrees in many respects with the one given by Kantele *et al.* (1964). It should be noted that the 75-keV gamma transition is known (Sliv *et al.* 1956-58) to be highly converted ( $\alpha_T = 10.24$ ) and therefore in most of the experiments it was not observed. Instead a 50-keV Yb K X-ray appeared. In the decay scheme the gamma transitions of 350, 630, 870 and 1260 keV were assigned tentatively by Kantele *et al.* (1964). They are confirmed by coincidence measurements in our studies. The new gamma ray of 1350 keV is due to the transition from the 1880-keV level to the 525-keV level of the ground state rotational band in  $\text{Yb}^{174}$ . This is concluded from the coincidence results obtained by gating with the 50, 175 and 275-keV gamma



rays, with the 1200-1260 and 1200-1400-keV portions of the gamma spectrum (figs. 5 and 6) and also from the observed gamma spectrum in coincidence with the beta rays greater than 300 keV (fig. 7). From the studies of the Coulomb excitation of the rotational levels in  $Yb^{174}$ , it is known (De Boer *et al*, 1963) that the gamma cascade of energies 350-275-175-75 keV corresponds to the ground state rotational band  $K = 0$ . Assuming the 275, 175 and 75-keV as  $E2$  gamma transitions, the theoretically calculated total internal-conversion coefficients, ( $\alpha_T$ ) are 0.900, 0.424 and 10.24, respectively (Sliv and Band, 1956-58). By taking account of the  $\alpha_T$  values for these gamma transitions one obtains the following total intensities:  $99 \pm 15$ ,  $103 \pm 16$  and  $112 \pm 31$ , respectively, which are consistent with the decay scheme.

The ground state spin of the  $Tm^{174}$  can be assigned from the orbital systematics using Nilsson diagram. The 69th proton orbital  $1/2^+ [411 \downarrow]$  consistently appears in the ground state of Tm isotopes with mass numbers 165 to 173. Similarly, the 105th neutron orbital  $7/2^- [514 \downarrow]$  consistently appears in the ground state of  ${}_{70}Yb^{175}$ ,  ${}_{71}Lu^{176}$ ,  ${}_{72}Hf^{177}$ ,  ${}_{73}Ta^{178}$ , and  ${}_{74}W^{179}$ . Therefore one can reasonably assume that the ground state of  ${}_{69}Tm^{174}_{105}$  consists of these two configurations, and according to the coupling rules of Gallagher and Moszkowski (1958) the expected ground state spin of it should be  $4^-$ . The observed log ft values for the beta transitions from  $Tm^{174}$  are the allowed unhindered type and suggest that the spins of the 2380 and 1880-keV levels of  $Yb^{174}$  should be  $5^-$  or  $4^-$  or  $3^-$ . The

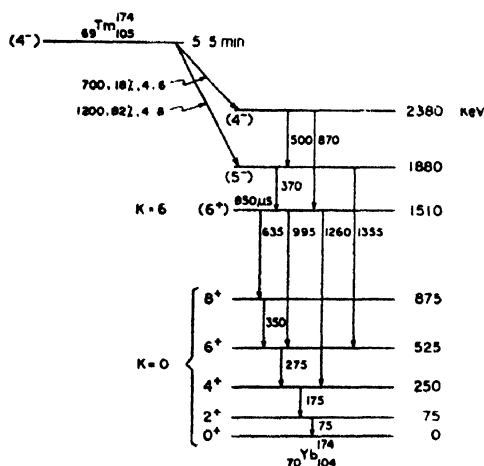


Fig. 8. Proposed decay scheme of  $Tm^{174}$ .

assignment of  $6^+$  (Katele, 1964) or probably  $7^-$  (Funke, *et al*, 1965) spin value to the 850- $\mu s$  isomeric state at 1510 keV and the observed gamma transitions from the levels at 2380 and 1880 keV to the 1510-keV state rule out the possibility of  $3^-$ . As suggested by Kantele *et al*, (1964) the pairing model calculations of Gallagher and Soloviev (1962) give the levels at 2300, 1800 and 1600 keV with the spins as  $4^-$ ,  $5^-$  and  $6^+$  which can fit with the observed levels at 2380, 1880

and 1510 keV, respectively. The  $4^-$  and  $5^-$  levels are the proton two-quasi-particle excitations having the configurations  $7/2 [523 \uparrow]$ ,  $1/2^+ [411 \downarrow]$  and  $1/2^+ [411 \downarrow]$ ,  $9/2^- [514 \uparrow]$ , respectively and the  $6^+$  level is due to the neutron two-quasi particle configuration  $5/2^- [512 \uparrow]$  and  $7/2^- [514 \downarrow]$ .

We have calculated the hindrance factors for the 1260, 990 and 630-keV gamma transitions from the observed intensities for different multipolarities obtained by assuming  $6^+$  and  $7^-$  spins for the 850- $\mu$ s isomeric state. It is found that  $6^+$  assignment to the 1510-keV isomeric state in  $\text{Yb}^{174}$  is more consistent with our observations instead of  $7^-$  arising due to the neutron two-quasi particle configurations  $9/2^+ [624 \uparrow]$ ,  $5/2^- [512 \uparrow]$ . From the  $(\text{O}^{16}; \text{O}^{16}\gamma)$  reaction on  $\text{Yb}^{174}$ , the levels at 75, 250, 525 and 875 keV which form a group of the ground state rotational band  $K = 0$  are assigned the spins as  $2^+$ ,  $4^+$ ,  $6^+$  and  $8^+$ , respectively (De Boer *et al.*, 1963).

#### ACKNOWLEDGMENTS

The authors wish to express their gratitude to Professor B. D. Nagchaudhuri and Professor D. N. Kundu for their kind interest. They are also indebted to Dr. H. Bakhru and Mr. B. Sethi for the helpful discussions and to the technical staff of the Neutron Physics Section for sample irradiations.

#### REFERENCES

- Chatterjee, A., 1964, *Nuclear Phys.*, **60**, 273.  
 De Boer, J., Goldring, G. and Winkler, H., 1963, *Proc. Third Conf. on Reactions between Complex Nuclei*, Asilomar, Calif., 317.  
 Funke, L., Graber, H., Kaun, K. H., Sodan, H., and Weruer, L. 1965, *Nuclear Phys.*, **61**, 465.  
 Gallagher, Jr., C. J. and Moszkowski, S. A., 1958, *Phys. Rev.*, **111**, 1082.  
 Gallagher, Jr., C. J. and Soloviev, V. G., 1962, *Mat. Fys. Skr. Dan. Vid. Selsk.*, **2**, No. 2.  
 Gardner, D. G., 1962, *Nuclear Phys.*, **29**, 373.  
 Gujrathi, S. C. and Mukherjee, S. K., 1965, (to be published).  
 Kantele, J., Broom, K. M. and Chittenden II, D. M., 1964, *Ann. Acad. Fenni. Series A* **VI**, No. 162.  
 Kantele, J., 1964, *Phys. Lett.*, **11**, 59.  
 Mukherjee, S. K., Ganguli, A. K. and Majumdar, N. K., 1961, *Proc. Phys. Soc. (London)*, **77**, 508.  
 Nuclear Data Sheets, 1964, *National Academy of Sciences*, National Research Council, Washington, D.C.  
 Orth, C. J., 1964, *Bull. Am. Phys. Soc.*, **9**, 498.  
 Slater, D. N., 1962, *Gamma rays of radio-nuclides in order of increasing energies*, Butterworths Scientific Publications (London).  
 Sliv, L. A. and Band, M. I., 1956-58, *Tables of internal conversion coefficients, of gamma rays*, Academy of Sciences, Press USSR, Moscow-Leningrad.  
 Takahasi, K. Kuroyangi, H. Y., Kotajima, K., Nagatani, K., and Morinaga, H., 1961, *J. Phys. Soc. (Japan)*, **16**, 1664.  
 Wille, R. G. and Fink, R. W., 1960, *Phys. Rev.*, **118**, 242.